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USING METHYL IODIDE

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NONRADIOMETRIC AND RADIOMETRIC TESTING OF RADIOIODINE SORBENTS USING METHYL IODIDE*

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Abstract

A nonradiometric test of adsorbents and adsorbers with normal methyl iodide ($\text{CH}_3 \text{}^{127}\text{I}$) is desirable. Use of methyl radioiodide ($\text{CH}_3 \text{}^{131}\text{I}$) requires special precautions and facilities and results in bed contamination. However, first it must be established to what extent the removal of $\text{CH}_3 \text{}^{127}\text{I}$ by adsorbents is indicative of the removal of $\text{CH}_3 \text{}^{131}\text{I}$.

An experimental apparatus was built and used to simultaneously measure the penetrations of CH_3I molecules and the radioisotope in $\text{CH}_3 \text{}^{131}\text{I}$ through charcoal adsorbent beds. Gas chromatography with electron capture detection was used to measure CH_3I . Radioiodine was measured using charcoal traps within NaI scintillation well crystals. Real time (5-min interval) radioiodine measurement provided immediate penetration results directly comparable to the real time penetrations of methyl iodide.

These penetrations were compared for typical charcoal adsorbents with these impregnants: a) 5% KI_3 , b) 5% KI_3 + 2% TEDA, c) 5% TEDA, and d) metal salts (Whetlerite). Differences between CH_3I and $\text{CH}_3 \text{}^{131}\text{I}$ penetrations observed for the two iodized charcoals were attributed to isotope exchange reactions. Equivalent penetrations were observed for non-iodized adsorbents and for iodized ones at initial time. First order rates were confirmed for reactions with TEDA and for isotope exchange. This was one more confirmation of the lack of a challenge concentration effect on efficiencies at low test bed loadings. In addition to other removal mechanisms, reversible physical adsorption was observed with all charcoals.

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1. Introduction

Testing of radioiodine adsorbents and adsorbers with nonradiometric vapors has several advantages. The test agents have lower toxicity, requiring fewer handling precautions, fewer special facilities, and no licensing for field use. Their use allows more frequent tests due to lower cost of the test agent. There is no radioactive contamination of the test bed and no release of radioactive vapor in the bed effluent.

One such nonradiometric test in current use is the in-place leak test of adsorber stages using a refrigerant tracer gas (Freon) and one or two gas chromatographs with electron capture detectors.⁽¹⁾ This procedure gives penetrations which are extrapolated to zero test time. An unusually high initial penetration value ($>0.05\%$) indicates the need to locate and repair a leak and retest. Another example is the carbon tetrachloride activity test performed on activated carbon base materials to determine their surface areas and degrees of activation.⁽²⁾

Every compound has its distinctive adsorption or reactive behavior with a given adsorbent. Therefore, the most useful tests of removal efficiency involve the same vapor to be trapped. Radioiodine (^{131}I) tagged elemental iodine (I_2) and methyl iodide (CH_3I) are used for laboratory measurements of adsorbent efficiencies and retentions^(3,4) and for in-place testing of adsorber banks.⁽⁵⁾ These radioiodine species are the major vapor forms of ^{131}I identified in nuclear environments. Very sensitive measurements due to the high specific activity of ^{131}I allow determinations of small penetrations using small amounts of radioiodine vapors. A total efficiency value is obtained by an established test procedure involving bed humidification, loading, and elution for predetermined periods and conditions.

From time-to-time the possibility of using normal iodine (^{127}I) vapor species for efficiency testing has been proposed. Two papers at the 15th DOE Nuclear Air Cleaning Conference reported preliminary efforts at defining the usefulness of normal methyl iodide for this application. One paper by Wood, et al., showed experimental breakthrough curves due to methyl iodide loading on several types of charcoal adsorbents.⁽⁶⁾ Effects of relative humidity, challenge vapor concentration, and linear velocity of air on breakthrough times were demonstrated. In retrospect, the breakthrough times were measures of bed capacities, rather than of efficiencies that are more appropriate to the negligible bed loadings usually expected for radioiodine. However, these results did point out that the total methyl iodide loading ($\text{CH}_3\ ^{131}\text{I} + \text{CH}_3\ ^{127}\text{I}$) and not just the loadings of $\text{CH}_3\ ^{131}\text{I}$ can affect the efficiency of an adsorbent.

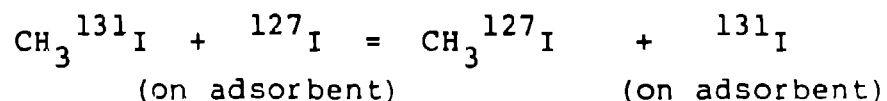
The paper by Romans and Dietz⁽⁷⁾ compared $\text{CH}_3\ ^{131}\text{I}$ and $\text{CH}_3\ ^{127}\text{I}$ penetrations using conditions representative of those in the RDT M16-1T standard test procedure. Good agreement was found between 2-h accumulated penetrations determined using $\text{CH}_3\ ^{127}\text{I}$

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and $\text{CH}_3^{131}\text{I}$ in separate tests. Penetrations agreed within a factor of 3 for a charcoal impregnated with KI_x and within a factor of 4 for a charcoal impregnated with $\text{KIO}_3 + \text{KI} + \text{K}_3\text{PO}_4 + \text{HMTA}$ (hexamethylenetetramine). A more detailed report of this work has been published recently.⁽⁸⁾

A recent paper by Jonas, Dietz, and Romans contained additional data on a charcoal with the latter set of four impregnants.⁽⁹⁾ Five tests with methyl radioiodide at three laboratories for 95% relative humidity and 16 h prehumidification resulted in an average penetration of 0.26% with a standard deviation of 0.16%. Measurements were again made under RDT M16-1T specifications. This average corresponds to a first order adsorption rate constant of 24 s^{-1} . In the one experiment using normal methyl iodide under similar conditions (16 h prehumidification at 90% relative humidity) an adsorption rate constant of 28 s^{-1} (1693 min^{-1}) was reported.

The agreements in these penetration and rate constant results are encouraging for using normal methyl iodide to indicate efficiency for methyl radioiodide. However, on deeper reflection, they are also puzzling. These adsorbents contain normal iodine in various forms to remove radioiodine by isotope exchange:



The normal methyl iodide formed is either trapped by another mechanism or passes out of the bed. Only if the isotope exchange rate is insignificant would equivalent removal efficiencies be expected for
 $\text{CH}_3^{131}\text{I}$ and $\text{CH}_3^{127}\text{I}$.

In an attempt to resolve this problem we have made extensive comparisons of $\text{CH}_3^{131}\text{I}$ and $\text{CH}_3^{127}\text{I}$ penetrations through test beds of four variously impregnated charcoals. The basic question addressed was: to what extent is the removal of methyl iodide by an impregnated charcoal indicative of the removal of methyl radioiodide?

II. Experimental Details

An experimental apparatus was constructed for periodic and simultaneous measurements of penetrations of both total methyl iodide (CH_3I) and methyl radioiodide ($\text{CH}_3^{131}\text{I}$) through test beds. Its components and design are shown in Figure 1. Compressed air was cleaned, filtered, regulated, and humidified before entering a 2.4-cm-i.d. glass flow system inside a glove box. An electronic mass flow meter (Datametrics 800-L) which monitored airflow was periodically checked using a dry test meter (Singer DTM-325) at the test bed location. Humidification was accomplished by passing air through the headspace over a heated water reservoir. A humidity monitor/controller (Phys-Chemical Research Corp.), which controlled water heating, was calibrated with a dew point hygrometer (EG&G 911) at the test bed location.

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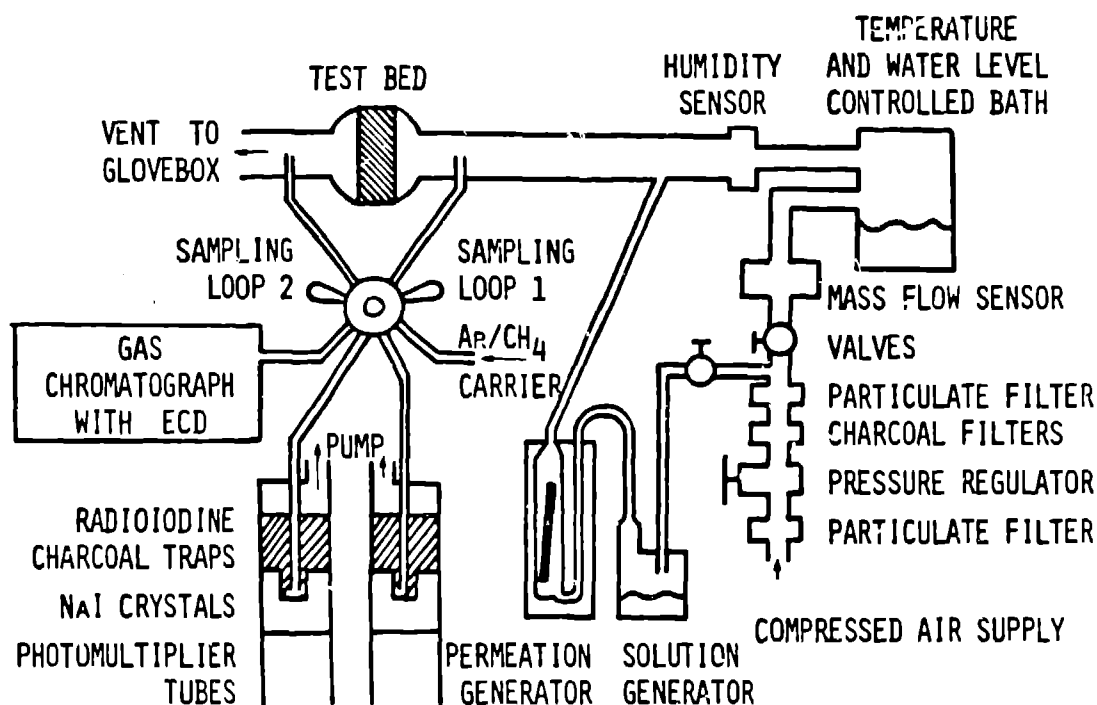


FIGURE 1

APPARATUS FOR SIMULTANEOUS CH_3I AND $\text{CH}_3^{131}\text{I}$ BED PENETRATION MEASUREMENTS

$\text{CH}_3^{127}\text{I}$ + $\text{CH}_3^{131}\text{I}$ Vapor Generation

Vapors for challenging test beds were generated in two ways: (1) Liquid methyl iodide containing $\text{CH}_3^{131}\text{I}$ was released at a steady rate by permeation through the walls of a sealed Teflon tube into a $500 \text{ cm}^3/\text{min}$ airflow. Permeation rate was regulated by temperature control ($25 - 76^\circ\text{C}$, $\pm 0.1^\circ\text{C}$) of the permeation tube in a Calibration System (Analytical Instrument Development, Inc., Model 303). The methyl iodide-radioiodide was ordered as $5\text{mCi }^{131}\text{I}$ in 3 mL total CH_3I (ICN Chemical and Radioisotope Division, Irvine, CA). Two milliliters were used to fill a permeation tube. (2) Alternately, methyl iodide containing $\text{CH}_3^{131}\text{I}$ was generated from an aqueous solution. A syringe was used to inject 2 mL of solution (0.1% CH_3I in distilled water) into 100 mL of water in a glass container. The volatile methyl iodides in the stirred solution entered the headspace and were swept by $500 \text{ cm}^3/\text{min}$ of air into the

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main airflow. These two generation methods represent two different situations in which radioiodine adsorbers are used. The steady, low level release of CH_3I from a permeation tube is typical of routine air cleaning situations. Generation from solution produces a spike of CH_3I concentration which drops exponentially with time, typical of an accidental radioiodine release.

Methyl Iodide Detectors

The detector for total methyl iodide was a gas chromatograph (Varian 1520) with a linearized ^{63}Ni electron capture detector (Tracor Instruments). Air from upstream and downstream of the test bed was drawn at $800\text{ cm}^3/\text{min}$ through matched Teflon sampling loops attached to a 10-port valve (Valco Instrument Co.) of Hastalloy-C (for inertness). This valve was pneumatically actuated by a digital valve sequence programmer (Valco Instrument Co.) to alternately inject the upstream and downstream air at 2.5-min intervals. The chromatographic column was 1.8-m x 4-mm-i.d. glass packed with 15% OV-7 on 100/120-mesh Chromosorb G. Operating conditions were 100°C and $20\text{ cm}^3/\text{min}$ 19:1 Ar: CH_4 carrier gas. An electronic peak integrator (Spectra Physics Minigrator) quantitated the methyl iodide peaks and recorded elapsed times. Calibrations used CH_3I permeation tubes whose outputs were determined by periodic weighings.

Methyl Radioiodide Detectors

The radiometric detectors collected and measured ^{131}I from the $800\text{ cm}^3/\text{min}$ air samples passing through the gas chromatograph sampling valve. Figure 1 shows the charcoal traps and 7.6-cm diam x 7.6-cm-thick NaI (Tl) well-type (52-mm deep x 29-mm-diam) scintillation crystals with integral photomultiplier tubes (Harshaw Chemical Co.). High-efficiency charcoal was used (5% TEDA impregnated Barnebey-Cheney CN 2762). The majority of radioiodine was collected at the bottom of the well, resulting in good detection efficiencies (~ 0.5) for the 0.364 MeV gamma-ray.⁽¹⁰⁾ Each detector for upstream and downstream air had its own preamplifier, amplifier, single-channel analyzer, and counter (all from Ortec). They shared the power bin (Ortec), high voltage power supply (Canberra), timer (Ortec), and printer (Ortec). Linear-log rate meters (Mech-Tronics) were used for count rate monitoring. Detector counts were taken for 5-min intervals and printed together. Each detector trap and crystal was shielded by 5 cm of lead to reduce background counts. Fresh charcoal was placed in the detector traps for background counts before each new bed was tested. The detectors were calibrated almost daily for relative sensitivities by sampling the same radioiodine-containing air.

Test Beds

Charcoal test beds of selected depths were packed into 2.4-cm-i.d. glass tubing. The usual airflow rate was 1.8 L/min, which corresponds to 6.6 cm/s linear velocity and 0.75 s residence time for a 5-cm-deep bed. This residence time is three times that specified in the standard radioiodine test method.⁽³⁾ However, this difference will not qualitatively affect the conclusions reached

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in this paper. Charcoals for testing were taken from air-purifying respirator canisters. Impregnations of the 8-16 mesh range charcoals were identified by the manufacturers (MSA = Mine Safety Appliances Company, Pittsburgh, PA; and Scott = Scott Health/Safety Products, South Haven, MI) as given in Table I.

Table I. Experimental Charcoals

<u>Canister</u>		<u>Charcoal Impregnants (Weight Percents)</u>
<u>Source</u>	<u>Designation</u>	
MSA	GMR-I	5% KI ₃ ^a
MSA	GMR-I ^b	5% KI ₃ + 2% TEDAC
MSA	GMR-S	Metal and Ammonium Salts ^d
Scott	600252-75	5% TEDA

^aKI₃ = equal amounts of KI and I₂

^bCanisters manufactured after July, 1979, through at least April 1980.

^cTEDA = triethylenediamine, N(CH₂CH₂)₃N

^dWhetlerite charcoal

III. Results: Iodized Charcoal without Amine Impregnant

Comparisons of CH₃I and CH₃¹³¹I Penetrations

To illustrate the type of results that were observed in 17 experiments with 5% KI₃-impregnated charcoal, we will first consider one test conducted under the following conditions:

3.75 cm-depth x 2.4-cm-diam bed
1.6 L/min airflow; 6.6 cm/s; 0.57 s bed contact time
86% RH; 2-h equilibration before testing
3.8 mg/m³ CH₃I; 0.125 µCi/m³ ¹³¹I

Cumulative CH₃ ¹³¹I penetrations after successive 5-min intervals were calculated from the ratios of downstream and upstream (of the test bed) decay counts, corrected for relative detector sensitivities. Instantaneous CH₃I penetrations at 5-min intervals were calculated from the ratios of peak areas obtained from downstream and upstream sampling, corrected for relative sampling loop volumes. For comparisons, cumulative CH₃I penetrations were calculated by integrating the instantaneous penetrations over time using a trapezoidal approximation. The results of these calculations are shown in Figure 2. The first observation is that cumulative CH₃I penetrations and cumulative CH₃ ¹³¹I penetrations were not equal except at initial experimental time (t=0). They diverged increasingly with increasing time. Secondly, instantaneous CH₃I penetration began to exceed 100% at 88 min due to desorption of previously adsorbed vapor. At this same time instantaneous CH₃ ¹³¹I penetration was only 11%.

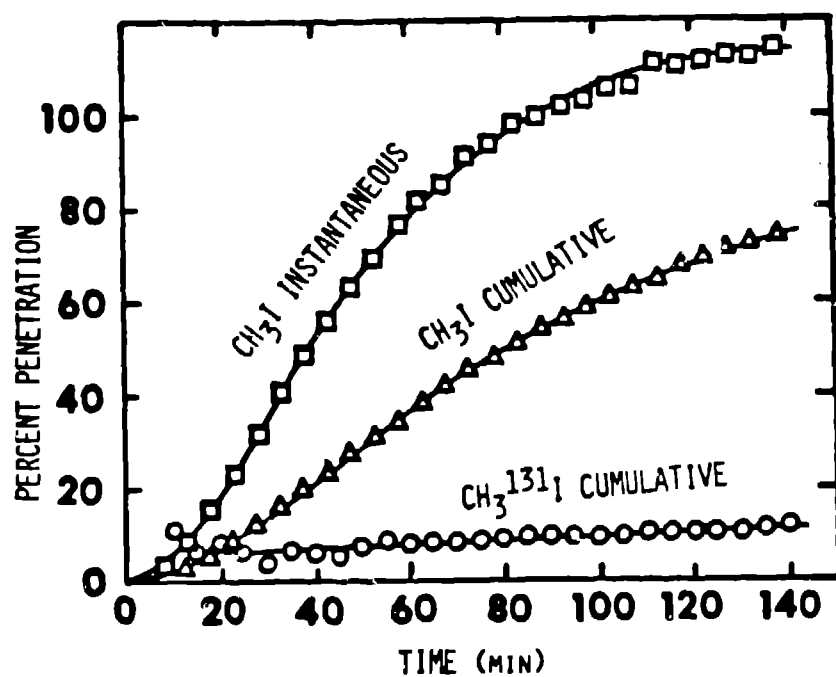


FIGURE 2
COMPARISONS OF PENETRATIONS FOR A TEST OF
AN IODIZED (ONLY) KI₃ CHARCOAL

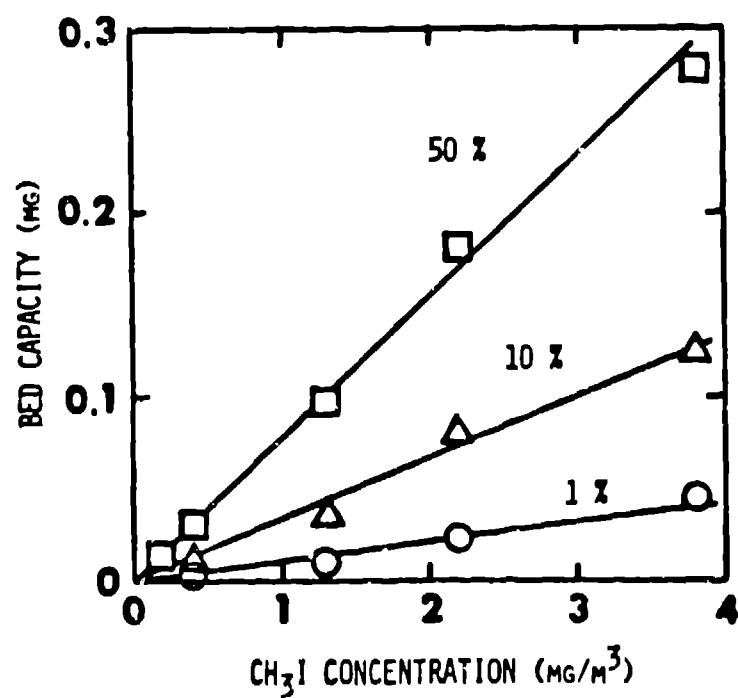


FIGURE 3
DYNAMIC ADSORPTION ISOTHERMS AT THREE INSTANTANEOUS PENETRATION
PERCENTS FOR METHYL IODIDE AND THE KI₃ CHARCOAL

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Effects of Challenge Concentrations

Five tests were made at the above conditions but with different challenge concentrations of CH_3I (0.19 - 3.78 mg/m^3) and $\text{CH}_3^{131}\text{I}$ (0.0061 - 0.125 $\mu\text{Ci}/\text{m}^3$) varying over a factor of 20. Breakthrough times (t_B) for 1%, 10%, and 50% instantaneous penetrations were nearly the same for all challenge concentrations (Table II). Individual breakthrough times were used to calculate the breakthrough capacities ($t_B C_0$) plotted vs. challenge concentrations in Figure 3. The linearity of these plots indicates that CH_3I adsorption and desorption occurred according to a simple linear isotherm (Henry's law). Other charcoals which have been tested with CH_3I have not indicated linear isotherms.

Table II. Effects of Challenge Concentrations

CH_3I				$\text{CH}_3^{131}\text{I}$	
Conc.	Breakthrough Times (min)			Conc.	Percent
(mg/m^3)	$t_{1\%}$	$t_{10\%}$	$t_{50\%}$	($\mu\text{Ci}/\text{m}^3$)	Penetration ^a
0.19	6.8	18.0	39.6	6.1	12.3
0.41	2.4	14.7	41.5	13.6	16.0
1.29	3.8	15.4	41.8	42.6	9.7
2.19	7.4	20.5	45.8	72.3	15.4
3.78	3.4	15.1	38.7	124.7	11.5
Average	4.8	16.7	41.5	Average	13.0
Std. Dev.	2.2	2.5	2.7	Std. Dev.	2.7

^aAverage instantaneous penetration after the initial period in which physical adsorption was significant.

The Wheeler adsorption equation predicts the logarithm of penetration as a linear function of time for low penetrations (< 15%), and such plots have been reported for CH_3I .⁽⁹⁾ Other published data do not show this semilog relationship.⁽⁶⁾ The penetration curves for the experiments reported here with the iodized charcoal consistently fit the Statistical Moments Theory (SMT) equations⁽⁶⁾ and an empirical exponential $C/C_0 = at^b$ equation better than the Wheeler equation. For example, four data sets from Figure 2 where penetrations were less than 15% yielded the correlations in Table III.

The consistent failure of the Wheeler equation to give the best fit of penetration results from many experiments brings to question its use in extrapolating to define initial penetration. However, it will always give a conservative (higher) initial value relative to the true one due to the curvature of the breakthrough curve. For this example, the Wheeler equation gave an initial penetration value of 0.33% (std dev = 0.18%) to be compared with the SMT initial value of 0.094%. One of the best fits of the breakthrough curves was for a $(C/C_0) = at^b$ empirical equation which has three difficulties: (1) It has no theoretical basis. (2) It does not allow extrapolation

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Table III. Fits of Penetration Data to Equations

2	Equation	Linear Correlation
		Coefficient, r
	Wheeler: $\ln(C/C_0) = \ln a + b t_B$	0.9256
	Exponential: $\ln(C/C_0) = \ln a + b \ln t_B$	0.9999
	SMT: $\frac{t_B^{-m_1+m_3/6m_2}}{x_B} = -\sqrt{m_2} + \frac{m_3}{6m_2} x_B$	0.9999

of penetration to initial test time. (3) Extrapolated values vary rapidly at short times; for this example, 0.020% at 0.5 min, 0.073% at 1 min, and 0.265% at 2 min.

Effects of Bed Depth and Contact Time

Another series of experiments was performed in which bed depth of the KI₃ charcoal was varied from 1.25 cm to 5.0 cm for these conditions:

2.4-cm-diam bed.
1.8 L/min airflow; 6.6 cm/s linear flow velocity
86% RH; 2-h equilibration before testing
3.3 mg/m³ CH₃I; 1.22 μ Ci/m³ ¹³¹I.

Ratios of cumulative CH₃I and CH₃ ¹³¹I penetrations are plotted vs. time in Figure 4. These ratios increase from unity with increasing time and are larger for deeper beds. The intercept of unity resulted from physical adsorption, which is the same for CH₃ ¹³¹I and CH₃ ¹²⁷I, being the most effective removal mechanism initially. As the bed became loaded and desorption became significant, isotope exchange removal of CH₃ ¹³¹I, but not CH₃ ¹²⁷I, became more important. Therefore, cumulative CH₃I penetration through an iodized charcoal such as this one, is not indicative of CH₃ ¹³¹I cumulative penetration after an arbitrary period.

Average CH₃ ¹³¹I penetrations were calculated for 17 experiments in which bed depths, concentrations, and linear flow velocities (6.6 - 13.3 cm/s) were varied. These are plotted vs. bed contact (t_c) time on a semilog graph in Figure 5. Bed contact time is bed depth divided by linear flow velocity. The results indicate a straight line with intercept of unity and a slope of $d \ln(C/C_0) / dt_c = -3.62 \text{ s}^{-1}$ (std dev = 0.27 s^{-1}). These results indicate that CH₃ ¹³¹I removal is by a simple first order reaction with a rate coefficient of 3.62 s^{-1} . Such kinetics are also confirmed by the lack of concentration dependence previously discussed above. The removal reaction must be isotope exchange, since total CH₃I is being removed much less efficiently.

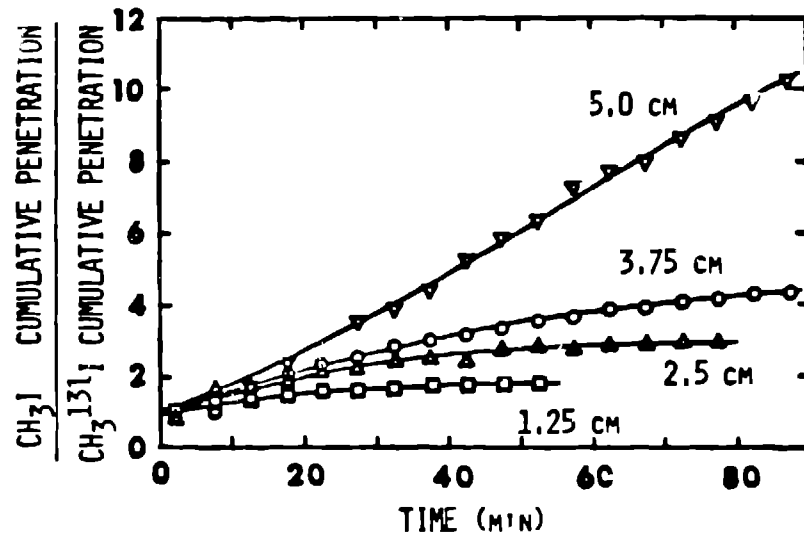


FIGURE 4
RATIOS OF $\text{CH}_3^{131}\text{I}$ AND CH_3I PENETRATIONS FOR FOUR DEPTHS
OF AN IODIZED (ONLY) KI_3 CHARCOAL

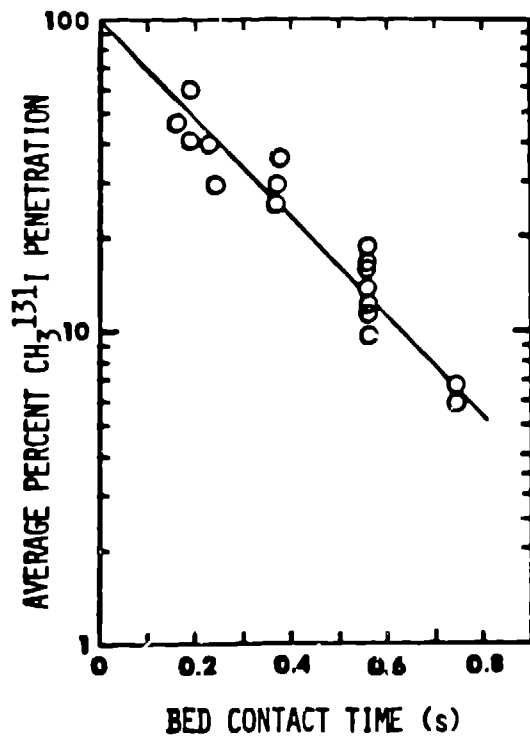


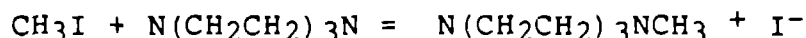
FIGURE 5
FIRST ORDER RATE PLOT FOR $\text{CH}_3^{131}\text{I}$
PENETRATION THROUGH IODIZED (ONLY)
 KI_3 CHARCOAL

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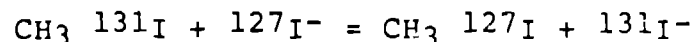
IV. Results: Iodized Charcoal with Amine Impregnation

Comparisons of CH₃I and CH₃¹³¹I Penetrations

Ratios of cumulative CH₃I and CH₃¹³¹I penetrations for three bed depths of charcoal with mixed impregnants (5% KI₃, 2% TEDA) are shown in Figure 6. Experimental conditions were similar to those of the preceding section. The initial equivalence of CH₃I and CH₃¹³¹I penetrations is again attributed to physical adsorption which is the same for both species. During the first 30 min, adsorption sites become exhausted and the most efficient removal mechanisms become:



for both species and



for methyl radioiodide only.

Effects of Bed Depth and Contact Time

Constant penetrations of CH₃I and CH₃¹³¹I after the initial physical adsorption period were averaged. Figure 7 shows semilog plots vs. residence time and bed depth (top scale). Again, straight lines with intercepts of unity resulted. The first order rate coefficient for total CH₃I removal by reaction with TEDA was 2.97 s⁻¹ (std dev = 0.12). The one for CH₃¹³¹I removal was 4.88 s⁻¹ (std dev = 0.15). The difference between these two values, 1.90 s⁻¹, must be the first order rate coefficient for isotope exchange.

V. Results: Amine (only) Impregnated Charcoal

Comparisons of CH₃I and CH₃¹³¹I Penetrations

Fourteen experiments were performed with the charcoal impregnated only with 5% TEDA. Bed depths (1.25 - 3.75 cm), relative humidities (75% or 86%), equilibration times (0 or 2 h), and generation methods (permeation tube or solution) were varied. Airflow velocity (6.6 cm/s) and bed diameter (2.4 cm) were constant. Challenge concentrations of CH₃I and CH₃¹³¹I varied widely, particularly during runs using vaporization from solution.

The penetrations of CH₃I and CH₃¹³¹I increased from zero to values constant with time. The initial period of efficient physical adsorption varied with relative humidity and period of bed equilibration prior to testing. Penetrations following the initial periods were averaged and compared in Figure 8. The plotted points

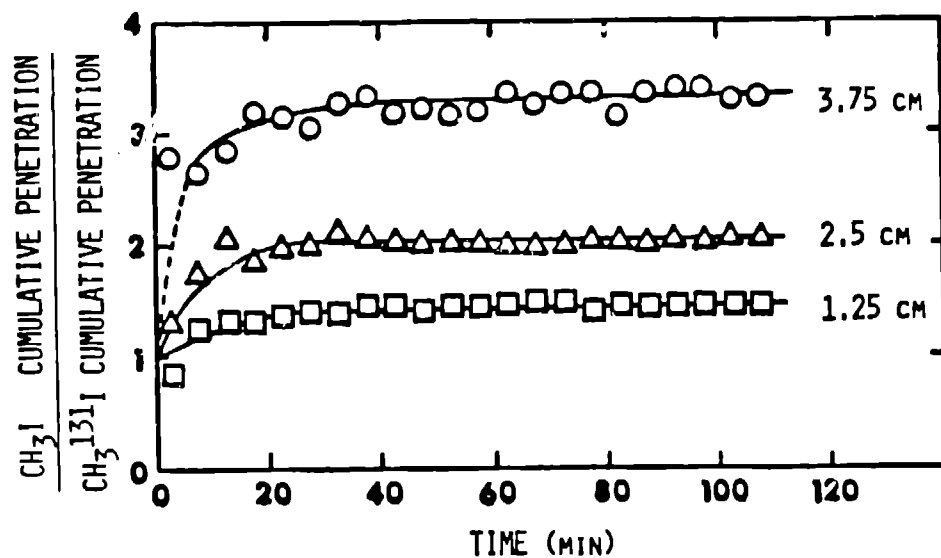


FIGURE 6
RATIOS OF $\text{CH}_3^{131}\text{I}$ AND CH_3I PENETRATIONS FOR THREE DEPTHS
OF A MIXED IMPREGNANT (KI_3 , TEDA) CHARCOAL

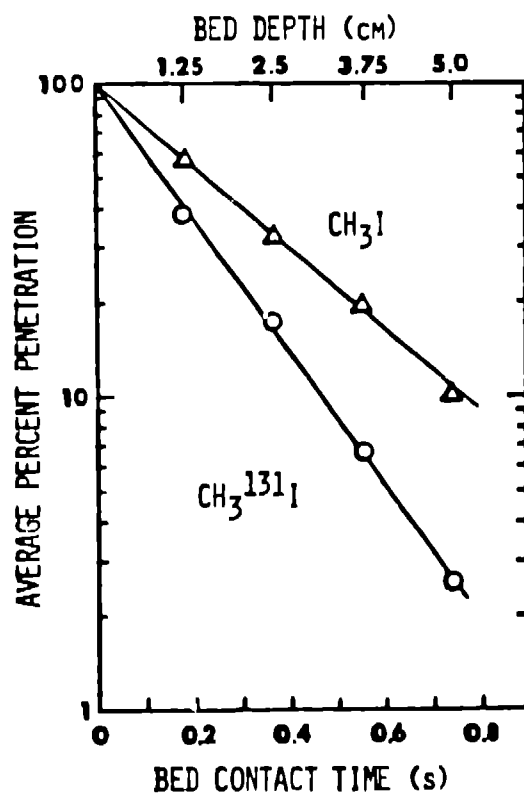


FIGURE 7
FIRST ORDER RATE PLOTS FOR $\text{CH}_3^{131}\text{I}$ AND CH_3I
PENETRATIONS THROUGH A MIXED IMPREGNANT
(KI_3 , TEDA) CHARCOAL

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all fall close to the dashed equality line. Therefore, as expected, CH_3I and $\text{CH}_3^{131}\text{I}$ were removed at equal efficiencies and rates by reaction with TEDA.

Effects of Bed Depth and Contact Time

Once again, plots of $\ln (C/C_0)$ vs t_c or bed depth for both CH_3I and $\text{CH}_3^{131}\text{I}$ were linear with intercepts of unity. Calculated rate coefficients are given in Table IV. Within experimental uncertainties there are no differences between any of these rate coefficients.

Table IV. First Order Rate Coefficients for Removal

<u>Species</u>	<u>Relative Humidity</u>	<u>Number of Points</u>	<u>Rate Coefficient (s^{-1})</u>	
			<u>Average</u>	<u>Std Dev</u>
CH_3I	86	10	7.10	0.30
$\text{CH}_3^{131}\text{I}$	86	10	6.91	0.46
Both	86	20	7.00	0.27
CH_3I	75	4	7.44	1.27
$\text{CH}_3^{131}\text{I}$	75	4	7.06	0.86
Both	75	8	7.25	1.03

VI. Results: Whetlerite Charcoal

Duplicate experiments were performed with beds of Whetlerite charcoal under conditions similar to those of Section III, A., but without equilibration before testing. Instantaneous penetrations of CH_3I increased with time and even exceeded 100% as the vapor adsorbed at the beginning was displaced. This is typical of removal by physical adsorption only. Breakthrough times averaged 33 ± 3 min at 0.1%, 49 ± 6 min at 1%, and 68 ± 8 min at 10% penetrations.

Cumulative penetrations of CH_3I and $\text{CH}_3^{131}\text{I}$ are compared in Figure 9. The data points close y fit the equivalence line until 100% penetration, then deviate slightly in the direction of greater $\text{CH}_3^{131}\text{I}$ penetration. This deviation is explained as the result of forming volatile radioiodides other than $\text{CH}_3^{131}\text{I}$. The ^{131}I detector is not compound specific, but the GC is. Indeed, other GC peaks were observed, but not identified or quantitated.

VII. Conclusions

A. Differences between CH_3I and $\text{CH}_3^{131}\text{I}$ removal rates were observed for two iodized (5% KI_3) charcoals. The charcoal also containing 2% TEDA had a lower first order isotope exchange rate coefficient (1.9 s^{-1}) than the one without (3.6 s^{-1}).

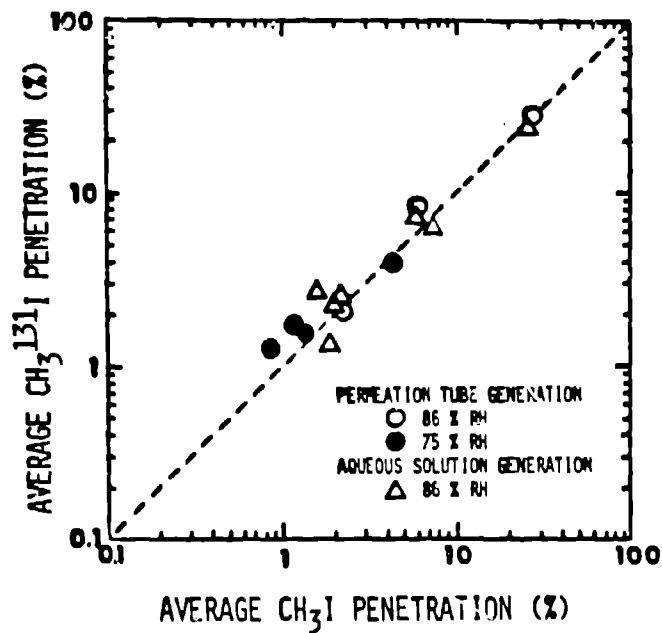


FIGURE 8
COMPARISONS OF $\text{CH}_3^{131}\text{I}$ AND CH_3I PENETRATIONS THROUGH BEDS
OF AMINE (ONLY) IMPREGNATED CHARCOAL

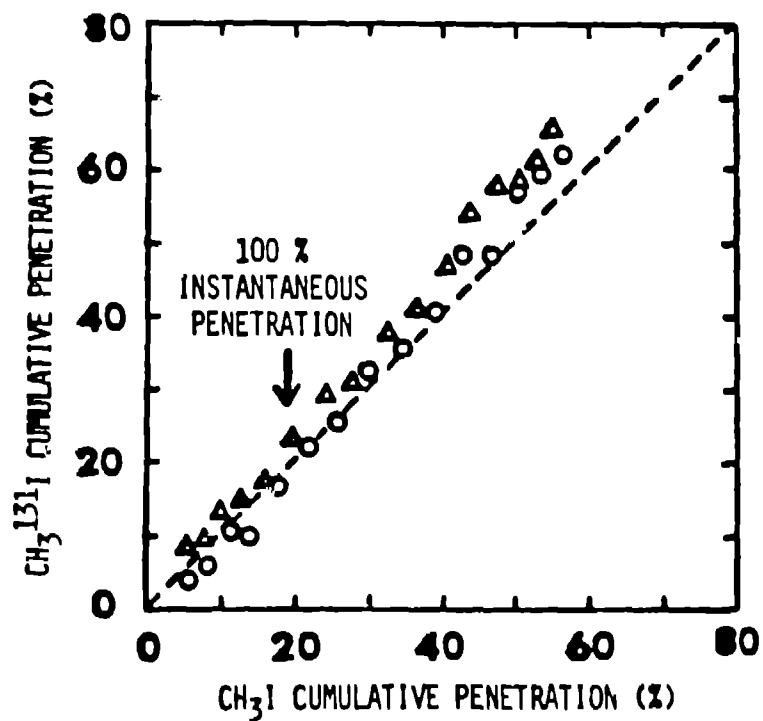


FIGURE 9
COMPARISONS OF $\text{CH}_3^{131}\text{I}$ AND CH_3I PENETRATIONS THROUGH BEDS
OF A WHETLERITE CHARCOAL

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B. Reactive trapping of CH_3I and $\text{CH}_3^{131}\text{I}$ was observed for two charcoals impregnated with TEDA. This was also by a first order rate mechanism with rate coefficients of 7.1 s^{-1} (5% TEDA) and 3.0 s^{-1} (2% TEDA + 5% KI_3).

C. Physical adsorption was also observed for all charcoals used. With KI_3 and/or TEDA impregnated charcoals, physical adsorption was more efficient than isotope exchange or reaction only at short times. Reversibility of physical adsorption results in cumulative penetrations approaching 100% unless these impregnants or similar ones are present.

D. CH_3I penetration is equivalent to $\text{CH}_3^{131}\text{I}$ penetration for

1. Iodized charcoals only at zero time;
2. Amine (only) impregnated charcoals at all times;
3. Charcoals without amine or ^{127}I impregnants at all times.

E. All experimental data indicated no effects of varying challenge concentrations on $\text{CH}_3^{131}\text{I}$ or CH_3I removal efficiencies by isotope exchange or TEDA reaction. The one iodized charcoal without amine showed no effect of challenge concentration on breakthrough times of CH_3I .

VIII. Applications of CH_3I as a Testing Agent

Laboratory Testing of Adsorbents

1. The efficiency of amine impregnation can be tested in the development of new sorbents, the acceptance testing of commercial ones, and the performance testing of used ones. Fewer precautions in handling and storage are required relative to the use of $\text{CH}_3^{131}\text{I}$. More rapid results are obtained by avoiding the need for decay counting.

2. Adsorption parameters for charcoals without amine impregnants can be determined. Methyl iodide can replace carbon tetrachloride in adsorbent qualification prior to impregnation.(2) Methyl iodide is more realistic and much less toxic than carbon tetrachloride. The development of the breakthrough curve can be used to determine efficiency and capacity for total CH_3I ($\text{CH}_3^{127}\text{I}$ + $\text{CH}_3^{131}\text{I}$ + etc.).

3. When tagged with ^{131}I , CH_3I can be used as described above to determine the efficiency of ^{127}I impregnation for isotope exchange.

In-Place Testing of Adsorbers

1. Leak testing of installed adsorbers can be done with CH_3I as well as with the Freons currently in use.(1) The problem of the best way to extrapolate penetration to zero time must be further studied.

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2. Total efficiency (adsorption plus desorption) testing of amine impregnation can be performed under actual use conditions of humidity, airflow rate, etc.

3. With iodized charcoals, as well as with amine impregnated charcoals, in-place CH_3I testing can be used to monitor the condition of adsorbers in use. Weathering and poisoning can be checked until the need for laboratory testing is indicated. This should reduce the frequency of the costly and difficult procedures of taking representative samples and laboratory testing.

X. References

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